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March 18, 2004

Re: 6335-97282/CN1113921

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Chinese Patent Publication No. 1113921A

Job No.: 6335-97282 Ref.: CN1113921
Translated from Chinese by the Ralph McElroy Translation Company
910 West Avenue, Austin, Texas 78701 USA

PEOPLE'S REPUBLIC OF CHINA
NATIONAL OFFICE OF INTELLECTUAL PROPERTY
INVENTION AND PATENT APPLICATION PUBLICATION JOURNAL
PUBLICATION NO. 1113921A

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|---|---|
| Int. Cl.[6]: | C08L 63/00<br>C08K 5/54<br>H01L 23/29 |
| Filing No.: | 94105911.1 |
| Filing Date: | June 2, 1994 |
| Publication Date: | December 27, 1995 |

# EPOXY RESIN COMPOSITION FOR PLASTIC SEALING OF SEMICONDUCTOR PARTS AND PREPARATION METHOD THEREOF

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Abstract

An epoxy resin composition, belonging to the field of resin compositions for packaging/sealing of semiconductors. The hydrolyzing medium for the silane coupling agent in said epoxy resin composition is an aqueous ammonia solution, and through regulating the acidity-alkalinity of the silane coupling agent solution, the reactivity of the silane coupling agent on the silicon dioxide powder surface is enhanced, which accelerates the formation of a homogeneous molecular monolayer of the silane coupling agent on the silicon dioxide powder surface.

Claims

1. An epoxy resin composition, characterized in that said epoxy resin composition has the following composition and blending ratio:

| Name of component | Parts by weight |
|---|---|
| 1. Epoxy resin | 100 |
| 2. Linear phenol formaldehyde resin | 50-70 |
| 3. Promoter | 0.5-15 |
| 4. Silicon dioxide micropowder treated with silane coupling agent in alkaline medium | 300-600 |

Said silane coupling agent is a low molecular weight compound having two different reactive functional groups in each molecule, for which the hydrolyzing medium is an alkaline solution of pH 8-13.

2. An epoxy resin compound according to the description in Claim 1, characterized in that said alkaline solution is an aqueous ammonia solution, for which the most optimal pH is 10-11.

3. A preparation method of an epoxy resin composition, characterized in that the hydrolyzing solution of said silane coupling agent is an alkaline solution of pH 8-13, and that the most optimal alkaline medium is an aqueous ammonia solution of pH 10-11.

Description

An epoxy resin composition for plastic packaging/sealing of semiconductor parts and preparation method thereof

This invention pertains to an epoxy resin composition for plastic packaging/sealing of semiconductor parts, particularly pertaining to an epoxy resin composition with improved adhesion and improved waterproofing property.

In general, silane coupling agents are used to treat inorganic fillers to improve the waterproofing property of a resin composition for tight sealing of semiconductors. A silane coupling agent is sprayed onto the surface of silicon dioxide by a dry method, but the drawback of said method is uneven spraying; in a wet method, silicon dioxide powder is dispersed in water or a solvent by high speed agitation, followed by adding the silane coupling agent, and although in said method the silane coupling agent and the silicon dioxide can make full contact, there is insufficient reactivity, which leads to an unsatisfactory treatment effect and low strength of the resultant composition; in particular, the bending strength is only about 1000 $kg/cm^2$.

To overcome the above drawbacks, Japanese Sumitomo Bakelite Plastic K.K. in 1988 disclosed in a patent (JP1-242658) in which silicon dioxide was pulverized first, followed by

adding a silane coupling agent, but the result was still not satisfactory because there was still uneven contact between the silane coupling agent and silicon dioxide powder.

Japan Fujitsu K.K in 1986 disclosed in a patent application (JP62-212420) that inorganic fillers be coated with a coupling agent containing an aminosilane but in order to have sufficient coating of the coupling agent, the amount of coupling agent had to be increased, but increasing the amount of coupling agent resulted in bad fluidity during molding.

Toshiba K.K in 1989 (JP2-218735), Matsushita Denko K.K. in 1988 (JP2-124923), Matsushita Denko K.K. in 1988 (JP2-86648) and Nitto Denko K.K. in 1989 (JP3-68158) respectively disclosed improvements in adhesion and thermal resistance by treating silicon dioxide powder using various silane coupling agents containing an amino group, an amino group [sic] or an epoxy group.

To improve the waterproofing property of the sealing resin, Nitto Denko K.K. in 1990 (JP4-38856) disclosed an inorganic filler system comprising silicon dioxide of 20-70 μm (X) and silicon dioxide powder of mean particle size of 0.1-10 μm (Y) treated by coating with an aminosilane coupling agent.

In the aforementioned conventional techniques, the silane treatment agent showed uneven coating on the surface of silicon powder after the inorganic filler was treated with the silane treatment agent.

To improve the waterproofing property, anti-bending property and adhesion, the present invention overcomes the uneven treatment of silicon dioxide powder with a silane coupling agent in the conventional techniques by providing an epoxy resin composition, and through adjusting the acidity-alkalinity of the silane coupling agent, the reactivity of the silane coupling agent on the surface of the silicon dioxide powder is enhanced and an even molecular monolayer of the silane coupling agent is rapidly formed on the silicon dioxide powder surface.

The epoxy resin composition of the present invention has the following composition and blending ratio:

| Name of component | Part by weight |
|---|---|
| 1. Epoxy resin | 100 |
| 2. Linear phenol formaldehyde resin | 50-70 |
| 3. Solidifying promoter | 0.5-15 |
| 4. Silicon dioxide micropowder treated with silane coupling agent in alkaline medium | 300-600 |

The aforementioned epoxy may be:

An ortho-cresol formaldehyde type epoxy resin, phenol formaldehyde type epoxy resin, bisphenol A type epoxy resin or fatty epoxy resin. The melting points of these resins are higher than room temperature, and these resins are either in solid form or in solution form with high

viscosity at room temperature. Said solidifying promoters are phenol formaldehyde resin, cresol formaldehyde resin, acid anhydrides and isocyanates.

The silicon dioxide powder is 400-mesh and 600-mesh silicon dioxide micropowder treated with a silane coupling agent in an alkaline medium.

Silane coupling agents are:

| Molecular formula | Name |
|---|---|
| 1. $H_2NCH_2CH_2CH_2Si(OC_2H_5)_3$ | γ-Aminopropyltriethoxysilane (compound A) |
| 2. $\underset{\diagdown O \diagup}{CH_2\text{-}CH}\text{-}CH_2\text{-}O(CH_2)_3\text{-}Si(OCH_3)_3$ | Propoxymethylpropylether trimethoxysilane (compound B) |
| 3. $HSCH_2CH_2CH_2Si(OCH_3)_3$ | γ-Thioaminopropyltriethoxysilane [sic; γ-Thiopropyltrimethoxysilane] |
| 4. $C_6H_5NHCH_2Si(OC_2H_5)_3$ | Phenylaminomethylethoxysilane [sic; Phenylaminomethyltriethoxysilane] |

The utilized silane coupling agent is a low molecular weight compound having two different reactive functional groups R and R' in the same molecule, and the general formula is:

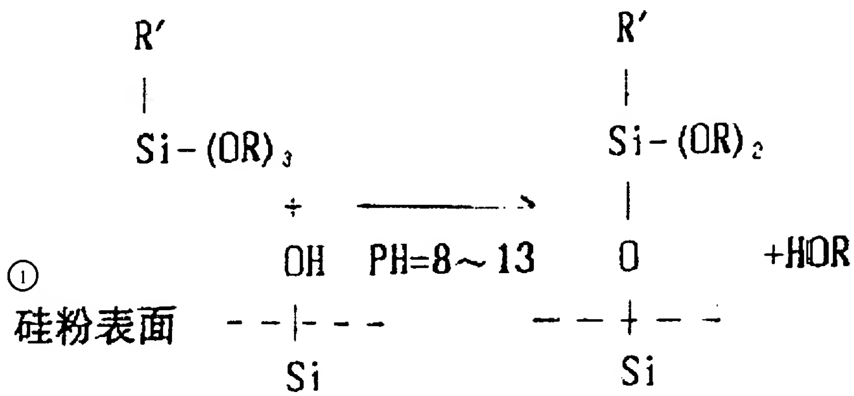
$$\begin{array}{l} R' \\ | \\ SI\text{-}(OR)_3 \end{array}$$

Where R' is $H_2NCH_2CH_2CH_2$—、 $\underset{\diagdown O \diagup}{CH_2\text{-}CH}\text{-}CH_2\text{-}O(CH_2)_3$—、 $HSCH_2CH_2CH_2$—

$C_6H_5NHCH_2$—

R is $CH_3$-, $C_2H_5$-

After hydrolysis, $R'\text{-}Si\text{-}(OR)_3$ undergoes chemical interaction with -OH groups on the surface of silicon dioxide micropowder to form Si-O-Si chemical bonds, which leads to the formation of a molecular monolayer coating of the silane coupling agent on the silicon powder surface. The reaction is shown in the following:




Key: 1 Silicon powder surface

The rate of hydrolysis of the silane coupling agent is an important factor in the reaction. To accelerate the hydrolysis of the silane coupling agent, the acidity-alkalinity of the coupling agent emulsion solution is adjusted by adding ammonia water to give a pH of 9-13 for the emulsion solution, and the most optimal pH is 10-11.

Various additives such as a flame retardant, colorant and mold discharge agent may be added to the epoxy resin of the present invention for packaging/sealing semiconductors so long as the resin property is not adversely affected.

The preparation of the epoxy resin composition is implemented by the following processes.

1. Preparation of alkaline emulsion solution of silane coupling agent

Silane coupling agent 50-100 g was added to 3-6 kg deionized water, and 50-100 mL ammonia water is added to the mixture to give an emulsion of pH=10-13.

2. 400-mesh or 600-mesh silicon dioxide 5-10 kg is added to the aforementioned emulsion, followed by agitating and then placing in a 70-120°C heating chamber for 3-5 h, to allow thorough impregnation of the treating agent into the silicon dioxide and to give a silicon dioxide micropowder evenly coated with the silane coupling agent.

3.

| | |
|---|---|
| Epoxy resin | 100 parts by weight |
| Solidifying agent | 40-60 parts by weight |
| Solidifying promoter | 1 part by weight |
| Silicon dioxide micropowder treated with silane coupling agent in alkaline medium | 300-600 parts by weight |

The substances are weighed according to the aforementioned blending ratio, followed by adding them one by one to a double-axial glue-melting machine preheated to

70-80°C and mixing until homogeneous. The mixture is then cooled and pulverized to a powder. The powder material is then made into a cake, which is preheated by high frequency [waves] and molded by a stepwise molding method to seal integrated circuits.

The epoxy resin composition of the present invention is utilized in plastic sealing of integrated circuits, and pressure cooking testing (PCT, 121°C, 2 atm) and temperature cycle testing (TCT, -50°C/5 min to 150°C/5 min) show that the epoxy resin composition of the present invention has a good waterproofing property and good crack resistance. The anti-bending strength is 1400-1500 kg/cm$^2$.

Application Example 1

132 mL deionized water was poured into a 500 mL Erlenmeyer flask, and 2-4 mL ammonia water was added to prepare a solution of pH=10-11. γ-Aminopropyltriethoxysilane treatment agent 2 mL was added and mixed to give an emulsified solution, and the emulsion was placed on a porcelain plate, and 200 g of 400-mesh or 200 g of 600-mesh silicon powder were added to the emulsion and the result was mixed until homogeneous. The porcelain plate was then placed in a 120°C heating chamber for 2 h, and after drying, the water content of the silicon powder was 0.2%-0.3%.

Application Example 2

3 kg deionized water was added to 50 kg propoxymethylpropylether trimethoxysilane, and 50 mL ammonia water were added to prepare a solution of pH=10 and 5 kg of 400-mesh silicon powder or 5 kg of 600-mesh silicon powder were added to the emulsion and the result was mixed until homogeneous, followed by heating in a 120°C heating chamber for 4.5 h, and after drying, the water content of the silicon powder was 0.2%-0.3%.

Ortho-cresol formaldehyde epoxy resin 100 parts, linear phenol formaldehyde 50 parts, solidifying promoter 1 part and 300 parts of silicon powder treated with propoxymethylpropylether trimethoxysilane in an ammonia medium at pH=10 were mixed and then further mixed at 80°C on a double-axial glue-melting machine, and the mixture was removed, dried and pulverized to a give powder material.

The powder material was pre-molded into a cake, which was preheated at high frequency and utilized for plastic sealing of integrated circuits by a stepwise molding method.

The static bending strength of said plastic sealing material was 1420 kg/cm$^2$, and Table II[*] shows the TCT and PCT results.

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[*] [Editor's note: Table II not in copy submitted for translation.]

The silicon micropowders utilized in Application Examples 3 and 4 were silicon powders treated by hydrolysis with a silane coupling agent in an ammonia water medium, and Tables I and II show the blending ratios and properties.

The silicon micropowders utilized in Comparison Examples 1 and 2 were silicon powders treated with a silane coupling agent in a neutral medium, and Tables I and II show the blending ratios and properties.

Table I. In parts by weight

| | ① 实施例 | | | ② 比较例 | |
|---|---|---|---|---|---|
| | 2 | 3 | 4 | 1 | 2 |
| ③ 环氧树脂 | 100 | 100 | 100 | 100 | 100 |
| ④ 线形酚醛树脂 | 50 | 50 | 50 | 50 | 50 |
| ⑤ 促进剂双腈氨 | 3 | 3 | 3 | 3 | 3 |
| ⑥ 硬脂酸脱膜剂 | 2. 5 | 2. 5 | 2. 5 | 2. 5 | 2. 5 |
| ⑦ 在PH=10时，硅烷偶联剂处理硅微粉 | ⑧ 偶联剂名称 | | | ⑨ 在PH=7时硅烷偶联剂处理硅微粉 | |
| | A | B | A | A | B |
| 400目 ⑩ | 200 | 150 | 100 | 200 | 150 |
| 600目 ⑩ | 200 | 250 | 300 | 200 | 250 |

Key:
1 Application example
2 Comparison example
3 Epoxy resin
4 Linear phenol formaldehyde resin
5 Promoter cyanoquanidine
6 Stearic acid mold discharge agent
7 Silicon micropowder treated with silane coupling agent at pH=10
8 Name of coupling agent

9 Silicon micropowder treated with silane coupling agent at pH=7
10 Mesh